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Apparatus and process for the preparation of caprolactam

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APPARATUS AND PROCESS FOR THE PREPARATION OF CAPROLACTAM

The invention relates to an apparatus for the preparation of caprolactam. The invention also relates to a process for preparing caprolactam.

Caprolactam can be prepared by Beckmann rearrangement of cyclohexanone oxime. Such Beckmann rearrangement can be carried out by admixing cyclohexanone oxime to a reaction mixture comprising caprolactam, sulfuric acid and optionally free SO_3. In such process the sulfuric acid and optional free SO_3 catalyse the conversion of cyclohexanone oxime towards caprolactam.

US-A-3,953,438 and US-A-3,914,217 disclose a more-stage rearrangement processes in which a first reaction mixture and a second reaction are kept in circulation, each mixture comprising caprolactam, sulfuric acid and free SO_3. Oleum and cyclohexanone oxime are fed into the first reaction mixture, while a portion of the first reaction mixture as well as cyclohexanone oxime are fed into the second reaction mixture. In a two-stage rearrangement process, caprolactam is recovered from a portion of the second reaction mixture that is withdrawn from the second circulating reaction mixture. In the three-stage rearrangement, a third reaction mixture comprising caprolactam, sulfuric acid and free SO_3 is kept in circulation, and a portion of the second rearrangement mixture and cyclohexanone oxime are fed into said third rearrangement mixture. Caprolactam is recovered from a portion of the third reaction mixture that is withdrawn from the third circulating reaction mixture. The more-stage rearrangement allows each stage to be operated at preferred values for the SO_3 concentration and M, M being, in the present application, defined as M defined as $(n_{SO3} + n_{H2SO4})/n_{cap}$, wherein n_{SO3} = quantity of SO_3 in reaction mixture, in mol, n_{H2SO4} = quantity of H_2SO_4 in reaction mixture, in mol, and n_{cap} = quantity of caprolactam in reaction mixture, in mol.

US-A-3,953,438 and US-A-3,914,217 also describe that sufficient mixing intensity should be applied for mixing the cyclohexanone oxime into the reaction mixture in order to obtain the lowest possible amount of impurities. Mixing is achieved using in-line mixers. It is also described that when a static mixer was used instead of an in-line mixer that the maximum amount of free SO_3 that could be tolerated was decreased, and that the purity decreased strongly (indicated by a rapid increase of the P.N. values) when using higher free SO_3 concentrations. Using the in-line mixers and

static mixers of US-A-3,953,438 and US-A-3,914,217, the mixing conditions cannot be selected per stage.

US-3,601,318 discloses a mixing device comprising closures. However, the mixing device is not used in a more-stage rearrangement.

We found that the yield of the process can be improved by selecting the mixing conditions per stage. Object of the invention is to provide an apparatus with which the mixing conditions can be selected per stage independently in a simple and effective way, such as to improve the yield.

This object is achieved by providing an apparatus for preparing caprolactam, said apparatus comprising:

a) a first circulation system for keeping a first reaction mixture in circulation, said first circulation system comprising a first mixing device for admixing cyclohexanone oxime to the first reaction mixture, said first mixing device comprising:
   (a1) a first tube through which the first reaction mixture can flow;
   (a2) first channels disposed around the first tube through which first channels cyclohexanone oxime can be fed into the first reaction mixture, said first channels opening into the first tube; and
   (a3) one or more first closures, one or more of the first channels being closable with a first closure; and
b) a second circulation system for keeping a second reaction mixture in circulation, said second circulation system comprising a second mixing device for admixing cyclohexanone oxime to the second reaction mixture, said second mixing device comprising:
   (b1) a second tube through which the second reaction mixture can flow;
   (b2) second channels disposed around the second tube through which second channels cyclohexanone oxime can be fed into the second reaction mixture, said second channels opening into the second tube; and
   (b3) one or more second closures, one or more of the second channels being closable with a second closure.

In a preferred embodiment, the apparatus further comprises.

c) a third circulation system for keeping a third reaction mixture in circulation, said third circulation system comprising a third mixing device for admixing cyclohexanone oxime to the third reaction mixture, said third mixing device comprising:
   (c1) a third tube through which the third reaction mixture can flow;

(c2) third channels disposed around the third tube through which third channels cyclohexanone oxime can be fed into the third reaction mixture, said third channels opening into the third tube; and

(c3) one or more third closures, one or more of the third channels being closable with a third closure.

The apparatus according to the invention comprises a first circulation system, a second circulation system, and preferably a third circulation system. The description of the preferred embodiments of the circulation system and preferred parts thereof refer to the preferred embodiments of the first circulation system and preferred parts thereof, the second circulation system and preferred parts thereof and the third circulation system and preferred parts thereof. A circulation system with which a reaction mixture can be kept in circulation may comprise any means with which at least part of the reaction mixture leaving the mixing device can be circulated back to the mixing device. The circulation system may comprise any connecting circuit through which the reaction mixture leaving the (tube of) the mixing device, can flow back to the (tube of) the mixing device. Preferably, the circulation system comprises a cooler for cooling the reaction mixture, and a connecting circuit through which the reaction mixture can flow from the mixing device to the cooler, and from the cooler back to the mixing device. Preferably, the circulation system comprises a pump for keeping the reaction mixture in circulation. Preferably, the pump is downstream of the mixing device and upstream of the cooler, as seen in the direction of flow of the reaction mixture. This facilitates achieving a high flow rate of the reaction mixture through the tube of the mixing device. An increased flow rate is found to result in an increased mixing intensity. Preferably, the circulation system comprises a collecting vessel arranged such as to receive the reaction mixture leaving the mixing device. Preferably, the collecting vessel is downstream of the mixing device (seen in the direction of flow of the reaction medium). Preferably, the pump is downstream of the collecting vessel and upstream of the cooler as seen in the direction of flow of the reaction medium.

The circulation system comprises a mixing device. The mixing device comprises a tube. Any suitable tube may be used through which a liquid may be passed. Preferably, the tube has a cylindrical shape. Preferably, the tube, as seen in the direction of flow, narrows, in a first part, to a throat and, optionally, widens beyond the throat in a second part. Preferably, the channels open into the first part, the throat or the second part of the tube, most preferably into the throat. As used herein the throat refers to the part of the tube beyond the first part (seen in the direction of flow) having

the smallest cross section area. The angle with which the first part narrows (angle between the wall of the first part and the axis of the tube) is preferably more than 5 °. The angle with which the second part narrows is widens is preferably more than 5 ° (angle between the wall of the second part and the axis of the tube).

According to the invention, channels are disposed around the tube. The channels may be any suitable openings through which cyclohexanone oxime can be fed into the reaction mixture. The channels may have any suitable diameter. The diameter of the channels is preferably at least 2 mm. This reduces the risk of clogging of the channels. The number of channels that are disposed around the tube may vary, and may for instance be between 2 and 32, preferably between 4 and 24. Preferably, the mixing device comprises a feed chamber, said feed chamber being disposed around the tube, from which feed chamber the channels open into the tube. The feed chamber may be connected to a source of cyclohexanone oxime, and cyclohexanone oxime may be fed from the feed chamber, through the channels into the tube.

According to the invention, the mixing device comprises one ore more closures, one or more of the channels being closable with a closure. Preferably each of the channels is closable with a closure. As closure can be used any suitable closure means with which a channel can be closed and opened, for instance a plug. Preferably a closure or plug is used having a tip complimentary in shape to the channel. Preferably, the closures or plugs have a tip complimentary in shape to the channels. This is an effective way of closing a channel.

In a preferred embodiment, the first tube extends through the wall of the first collecting vessel, the second tube extends through the wall of the second collecting vessel and/or the third tube extends through the wall of the third collecting vessel, such that the closures are still outside the collecting vessel. This facilitates the use of the closures.

Preferably, cyclohexanone oxime is filtered before admixing cyclohexanone oxime to the reaction mixture.

The apparatus may comprise any means for feeding oleum into the first reaction mixture. The first circulation system may comprise any suitable inlet for feeding oleum into the first reaction mixture. In a preferred embodiment, the first circulation system comprises an inlet for the oleum that is upstream of the pump of the first circulation system (seen in the direction of flow).

The apparatus may comprise any means for feeding a portion of the first reaction mixture into the second reaction mixture. The first circulation system may comprise any suitable outlet for the first reaction mixture. In a preferred embodiment, the first collecting vessel comprises an outlet, preferably an overflow, via which part of the first reaction mixture can be withdrawn. The second circulation system may comprise any suitable inlet for the first reaction that is withdrawn. In a preferred embodiment, the second circulation system comprises an inlet for the part of the first reaction mixture that is withdrawn, said inlet being upstream of the pump of the second circulation system (seen in the direction of flow of the second reaction mixture). This is an effective way of mixing the first reaction mixture into the second reaction mixture.

The apparatus may comprise any means for feeding a portion of the second reaction mixture into the third reaction mixture. The second circulation system may comprise any suitable outlet for the second reaction mixture. In a preferred embodiment, the second collecting vessel comprises outlet, preferably an overflow, via which part of the second reaction mixture can be withdrawn. The third circulation system may comprise any suitable inlet for the second reaction mixture that is withdrawn. In a preferred embodiment, the third circulation system comprises an inlet for the part of the second reaction mixture that is withdrawn, said inlet being upstream of the pump of the third circulation system (seen in the direction of flow of the second reaction mixture). This is an effective way of mixing the second reaction mixture to the third reaction mixture.

The invention also provides a process for preparing caprolactam using the apparatus according to the invention, said process comprising:

a) passing a first reaction mixture through the first tube, and keeping the first reaction mixture in circulation, said first reaction mixture comprising caprolactam, sulfuric acid and SO_3;
b) feeding cyclohexanone oxime into the first reaction mixture through the first openings.
c) passing a second reaction mixture through the second tube, and keeping the second reaction mixture in circulation, said second reaction mixture comprising caprolactam, sulfuric acid and free SO_3;
d) feeding into the second reaction mixture cyclohexanone oxime and a portion of the first reaction mixture, said cyclohexanone oxime being fed into the second reaction mixture through the second openings.

Preferably, the process further comprises:

e) passing a third reaction mixture through the third tube, and keeping the third reaction mixture in circulation, said third reaction mixture comprising caprolactam, sulfuric acid and free SO_3;

The first reaction mixture, the second reaction mixture, and the optional third reaction mixture comprise caprolactam, sulfuric acid and SO_3. The molar ratio M defined as $(n_{SO3} + n_{H2SO4})/n_{cap}$, wherein n_{SO3} = quantity of SO_3 in reaction mixture, in mol, n_{H2SO4} = quantity of H_2SO_4 in reaction mixture, in mol, and n_{cap} = quantity of caprolactam in reaction mixture, in mol, is preferably different in each reaction mixture. The molar ratio M in the first, second and third reaction mixture will, as used herein, be referred to as M(1), M(2) and M(3) respectively. The concentration SO_3 in the first, second, and third reaction mixture will, as used herein, be referred to as $C_{SO3}(1)$, $C_{SO3}(2)$ and $C_{SO3}(3)$. As used herein the SO_3 concentration will be given in wt.% relative to the weight of the reaction mixture. The temperature in the first, second and third reaction mixture will, as used herein, be referred to as T(1), T(2) and T(3) respectively. As used herein, the values for M, the SO_3 concentration, and the temperature refer in particular to the value in the reaction mixture obtained after feeding of the cyclohexanone oxime into the reaction mixture, in particular in the reaction mixture leaving the mixing device.

Preferred values for M and the SO_3 concentration can be obtained by feeding cyclohexanone oxime to the different stages in the appropriate amounts, and by applying appropriate quantities of oleum of appropriate SO_3 concentration.

Preferably, M(2) is lower than M(1). Preferably M(3) is lower than M(2).

In a preferred embodiment, M(1) is between 1.2 and 2.2, preferably between 1.4 and 1.85, more preferably between 1.5 and 1.7. Preferably, C_{SO3} (1) is between 3 and 20 wt.%, preferably higher than 4 wt.%, preferably higher than 6 wt.%, more preferably higher than 8 wt.%, more preferably higher than 10 wt.%, more preferably higher than 12 wt.%. Increased values for $C_{SO3}(1)$ have the advantage that $C_{SO3}(2)$ can be kept high in the second reaction mixture without having to feed oleum to the second reaction mixture. $C_{SO3}(1)$ may be less than 18 wt.%, preferably less than 17 wt.%. Preferably T(1) is between 50 and 130 °C, preferably between 70 and 130 °C, more preferably between 70 and 120 °C.

In a preferred embodiment M(2) is between 1.0 and 1.6, preferably higher than 1.1, more preferably higher than 1.2, preferably less than 1.5, more

preferably less than 1.4. Preferably, $C_{SO_3}(2)$ is between 0.5 and 20 wt.%, more preferably higher than 1 wt.%, more preferably higher than 2 wt.%, more preferably higher than 4 wt.%, more preferably higher than 6 wt.%, more preferably higher than 8 wt.%, more preferably higher than 10 wt.%, more preferably higher than 12 wt.%. Increased concentrations of $C_{SO3}(2)$ within the abovementioned ranges for M(2) were surprisingly found to result in significantly higher yields. Preferably T(2) is between 70 and 130 °C, preferably between 80 and 130 °C, more preferably between 80 and 120 °C.

In a preferred embodiment M(3) is between 1.0 and 1.4, preferably between 1.1 and 1.35, more preferably between 1.15 and 1.35. Preferably, $C_{SO3}(3)$ is between 0.5 and 18 wt.%, preferably higher than 1 wt.%, preferably higher than 2 wt.%, more preferably higher than 4 wt.%, preferably higher than 6 wt.%, more preferably higher than 8 wt.%, more preferably higher than 10 wt.%, more preferably higher than 12 wt.%. Increased concentrations of $C_{SO3}(2)$ within the abovementioned ranges for M(3) were surprisingly found to result in significantly higher yields. Preferably T(3) is between 70 and 130 °C, preferably between 80 and 130 °C, more preferably between 80 and 120 °C.

Oleum may be fed into a reaction mixture in any suitable way. Preferably all oleum applied is fed into the first reaction mixture. Preferably, the amount of cyclohexanone oxime fed to the first reaction mixture is larger than the amount of cyclohexanone oxime fed to the second reaction mixture, and, if applicable, preferably the amount of cyclohexanone oxime fed to the second reaction mixture is larger than the amount of cyclohexanone oxime fed to the third reaction mixture. Preferably, from 60 to 95 wt.% of the total amount of cyclohexanone oxime fed into the first, second and, if applicable, third reaction mixture, is fed into the first reaction mixture. Preferably, from 5 to 40 wt.% of the total amount of cyclohexanone oxime fed into the first, second and, if applicable, third reaction mixture is fed into the second reaction mixture. If applicable, preferably, from 2 to 15 wt.% of the total amount of cyclohexanone oxime fed into the first, second and third reaction mixture is fed into the third reaction mixture.

Preferably, one parts by volume of cyclohexanone oxime is continuously introduced into at least 10 parts by volume, more preferably at least 20 parts by volume of reaction mixture.

Preferably, $w1/W1 < 0.01$, preferably $w1/W1 < 0.05$. Preferably, $w2/W2 < 0.01$, preferably $w2/W2 < 0.05$, Preferably, $w3/W3 < 0.01$, preferably $w3/W3 <$

0.05, wherein w1, w2, w3 = flow rate (in m^3/s) of the cyclohexanone oxime which is fed through said one ore more first channels, second channels and third channels, respectively; and W1, W2, W3 = flow rate (in m^3/s) of the reaction mixture which is passed through the first tube, second tube and third tube respectively.

A continuous process according to the invention preferably involves feeding a portion of the first reaction mixture into the second reaction mixture. A continuous process according to the invention preferably involves withdrawing a portion of the second reaction mixture. A continuous process according to the invention may involve feeding a portion of the second reaction mixture into the third reaction mixture. A continuous process according to the invention may involve withdrawing a portion from the third reaction mixture.

A portion of the second reaction mixture and or of the third reaction mixture may be withdrawn in any suitable way. Caprolactam may be recovered from the second or third reaction mixture by known methods, for instance by neutralization with ammonia, and purification of the caprolactam-containing aqueous phase obtained.

We surprisingly found that the yield can be improved by selecting the number of first channels, second channels and/or third channels that are in closed position.

Preferably the process according to the invention is performed while a selected number of first channels, second channels and/or third channels is in closed position, said selected number being chosen such as to obtain a desired yield.

According to the invention cyclohexanone oxime is fed into the reaction mixture. The cyclohexanone oxime fed to the reaction mixture may comprise water, for instance less than 7 wt.%. Preferably, the cyclohexanone oxime fed into the reaction mixture has a water content of less than 2 wt.%, more preferably less than 1 wt.%, more preferably less than 0.2 wt.%, more preferably less than 0.1 wt.%. Feeding cyclohexanone oxime having a low water content is advantageous as it is an effective way for achieving a reaction mixture having high SO_3 content.

One way of obtaining cyclohexanone oxime having a water content of less than 2 wt.% is drying cyclohexanone oxime with a high water content for example with inert gas. A preferred way of obtaining cyclohexanone oxime having a water content of less than 2 wt.% is a process in which cyclohexanone oxime is obtained by

a) preparing an organic medium comprising cyclohexanone oxime dissolved in an organic solvent, and
b) separating, by distillation, cyclohexanone oxime from said organic medium.

Preparing an organic medium comprising cyclohexanone oxime dissolved in an organic solvent is preferably carried out by contacting in a reaction zone in countercurrent flow a stream of a solution of cyclohexanone in an organic solvent which is also a solvent for the cyclohexanone oxime and a stream of an a phosphate buffered, aqueous solution of hydroxylammonium; and withdrawing from the reaction zone an organic medium of cyclohexanone oxime dissolved in said organic solvent. Particularly suitable organic solvent for use in the process for preparing cyclohexanone oxime are toluene and benzene. Preferably toluene is used as organic solvent. The phosphate buffered, aqueous reaction medium is preferably continuously recycled between a hydroxylammonium synthesis zone and a cyclohexanone oxime synthesis zone. In the hydroxylammonium synthesis zone hydroxylammonium is formed by catalytic reduction of nitrate ions or nitric oxide with hydrogen. In the cyclohexanone oxime synthesis zone, hydroxylammonium formed in the hydroxylammonium synthesis zone reacts with cyclohexanone to form cyclohexanone oxime. The cyclohexanone oxime can then be separated from the aqueous reaction medium which is recycled to the hydroxylammonium synthesis zone. An organic medium comprising the formed cyclohexanone oxime dissolved in said organic solvent is withdrawn from the reaction zone, and distilled to recover cyclohexanone oxime having a water content less than 1 wt.% and even less than 0.1 wt.%.

Preferably, the process is performed while a selected number of first channels second channels, and/or third channels is in closed position, said selected number being chosen such as to obtain a desired v_1/V_1 and v_2/V_2 and optionally v_3/V_3, wherein

v_1 = the velocity (in m/s) at which cyclohexanone oxime is fed through the first channels into the first reaction mixture,

V_1 = velocity of the first reaction mixture at the level of said first channels in the first tube, V_1 being defined as W1/A1, wherein W is the flow rate (in m^3/s) of the reaction mixture that is fed into the tube and A1 is the cross section area of the tube (in m^2) at the level where said first channels open into the tube.

v_2 = the velocity (in m/s) at which cyclohexanone oxime is fed through the second channels into the second reaction mixture,

V_2 = velocity of the second reaction mixture at the level of said second channels in the second tube, V_2 being defined as W2/A2, wherein W2 is the flow rate (in m^3/s) of the second reaction mixture that is fed into the second tube and A2 is the cross section area of the second tube (in m^2) at the level where said second

channels open into the tube.

v_3 = the velocity (in m/s) at which cyclohexanone oxime is fed through the third channels into the third reaction mixture,

V_3 = velocity of the third reaction mixture at the level of said third channels in the third tube, V_3 being defined as W3/A3, wherein W3 is the flow rate (in m^3/s) of the third reaction mixture that is fed into the third tube and A3 is the cross section area of the third tube (in m^2) at the level where said third channels open into the tube.

In a preferred embodiment of the invention, v_1/V_1 is smaller than v_2/V_2, for instance $v_1/v_1 < 0.9*v_2/V_2$, for instance $v_1/V_1 < 0.8*v_2/V_2$, for instance $v_1/V_1 < 0.5*v_2/V_2$.

In another preferred embodiment of the invention, v_1/V_1 is higher than v_2/V_2, for instance $v_1/v_1 > 1.2*v_2/V_2$, for instance $v_1/V_1 > 1.5*v_2/V_2$, for instance $v_1/V_1 > 2*v_2/V_2$.

In another preferred embodiment of the invention, v_2/V_2 is smaller than v_3/V_3, for instance $v_2/V_2 < 0.9*v_3/V_3$, for instance $v_2/V_2 < 0.8*v_3/V_3$, for instance $v_2/V_2 < 0.5*v_3/V_3$.

In another preferred embodiment of the invention, v_2/V_2 is higher than v_3/V_3, for instance $v_2/V_2 > 1.2*v_3/V_3$, for instance $v_2/V_2 > 1.5*v_3/V_3$, for instance $v_2/V_2 > 2*v_3/V_3$.

In a preferred embodiment v_1/V_1 may be < 15, for instance < 10, for instance < 5, for instance < 2, for instance < 1.8, for instance < 1.5. The ratio v_1/V_1 may be > 0.2, for instance > 0.5.

In a preferred embodiment v_2/V_2 may be < 15, for instance < 10, for instance < 5, for instance < 2, for instance < 1.8, for instance < 1.5. The ratio v_2/V_2 may be > 0.2, for instance > 0.5.

In a preferred embodiment v_3/V_3 may be < 15, for instance < 10, for instance < 5, for instance < 2, for instance < 1.8, for instance < 1.5. The ratio v_3/V_3 may be > 0.2, for instance > 0.5.

<u>Description of preferred embodiment</u>

Figure 1 shows a preferred set-up for a rearrangement in three stages comprising a first circulation system, a second circulation system and a third circulation system. The first circulation system comprises mixing device A1, collecting vessel B1, pump C1 and cooler D1, and a first reaction mixture is kept in circulation via

line 1. The second circulation system comprises mixing device A2, collecting vessel B2, pump C2 and cooler D2, and a second reaction mixture is kept in circulation via line 11. The third circulation system comprises mixing device A3, collecting vessel B3, pump C3 and cooler D3, and a third reaction mixture is kept in circulation via line 21. Cyclohexanone oxime and oleum are fed into the first reaction mixture via line 2 and line 3 respectively. A portion of the first reaction mixture is withdrawn from collecting vessel B1 via line 4 and fed into the second reaction mixture. Cylcohexanone oxime is fed into the second reaction mixture via line 12. A portion of the second reaction mixture is withdrawn from collecting vessel B2 via line 14 and fed into the third reaction mixture. Cyclohexanone oxime is fed to the third reaction mixture via line 24. A portion of the third reaction mixture is withdrawn from collecting vessel B3 via line 34. The process is carried out continuously.

Figure 2 shows a mixing device that is preferably used as mixing device A1, mixing device A2, and mixing device A3. The mixing device comprises a cylindrical tube 101 that in first part 101a narrows to throat 101b, and beyond throat 101b widens in a second part 101c. The second part 101c of the tube is connected to a second tube 102. In the throat openings 103 are present which are in connection with feed chamber 104. The reaction mixture is fed through the tube in the direction of the arrow. Cyclohexanone oxime is supplied via feed chamber 104, and fed into reaction mixture through openings 103. The mixing device comprises closures 105 with which openings 103 can be opened and closed independently. By selecting the number of openings that are in closed position, the velocity of the cyclohexanone oxime can be selected for a given flow rate of the cyclohexanone oxime. (The mixing device also comprises a baffle 106 opposite to the exit of tube 101.) The tube opens into collecting vessel B, having walls 110, overflow 111, and outlet 112. Reaction mixture leaving tube 102 is collected in the collecting vessel B, and leaves collecting vessel B partly via line 112 to be further circulated, and partly via overflow 111 to be fed into a subsequent reaction mixture or for the recovery of caprolactam.

CLAIMS

1. Apparatus for preparing caprolactam, said apparatus comprising:
   a) a first circulation system for keeping a first reaction mixture in circulation, said first circulation system comprising a first mixing device for admixing cyclohexanone oxime to the first reaction mixture, said first mixing device comprising:
      (a1) a first tube through which the first reaction mixture can flow;
      (a2) first channels disposed around the first tube through which first channels cyclohexanone oxime can be fed into the first reaction mixture, said first channels opening into the first tube; and
      (a3) one or more first closures, one or more of the first channels being closable with a first closure; and
   b) a second circulation system for keeping a second reaction mixture in circulation, said second circulation system comprising a second mixing device for admixing cyclohexanone oxime to the second reaction mixture, said second mixing device comprising:
      (b1) a second tube through which the second reaction mixture can flow;
      (b2) second channels disposed around the second tube through which second channels cyclohexanone oxime can be fed into the second reaction mixture, said second channels opening into the second tube; and
      (b3) one or more second closures, one or more of the second channels being closable with a second closure.
2. Apparatus according to claim 1, wherein the apparatus further comprises:
   c) a third circulation system for keeping a third reaction mixture in circulation, said third circulation system comprising a third mixing device for admixing cyclohexanone oxime to the third reaction mixture, said third mixing device comprising:
      (c1) a third tube through which the third reaction mixture can flow;
      (c2) third channels disposed around the third tube, through which third channels cyclohexanone oxime can be fed into the third reaction mixture, said third channels opening into the third tube; and
      (c3) one or more third closures, one or more of the third channels being closable with a third closure.

3. Apparatus according to claim 1 or claim 2, wherein the first tube, second tube and/or third tube, as seen in the direction of flow, narrows, in a first part, to a throat, and, optionally, widens beyond the throat in a second part.

4. Apparatus according to any one of claims 1 to 3, wherein the first circulation system, the second circulation system and/or third circulation system comprises a collecting vessel, a cooler for cooling the reaction mixture, a connecting circuit through which the reaction mixture can flow from the mixing device to the collecting vessel, from the collecting vessel to the cooler, and from the cooler back to the mixing device.

5. Apparatus according to claim 4, wherein the first circulation system, the second circulation system and/or the third circulation system comprises a pump, said pump being downstream of the collecting vessel and upstream of the cooler as seen in the direction of flow of the reaction mixture.

6. Apparatus according to claim 4 or claim 5, wherein the first tube extends through the wall of the first collecting vessel, the second tube extends through the wall of the second collecting vessel and/or the third tube extends through the wall of the third collecting vessel, such that the closures are still outside the collecting vessel.

7. Process for preparing caprolactam using the apparatus according to any one of claims 1 to 6, said process comprising:

a) passing a first reaction mixture through the first tube, and keeping the first reaction mixture in circulation, said first reaction mixture comprising caprolactam, sulfuric acid and SO_3;

b) feeding cyclohexanone oxime into the first reaction mixture through the first openings.

c) passing a second reaction mixture through the second tube, and keeping the second reaction mixture in circulation, said second reaction mixture comprising caprolactam, sulfuric acid and free SO_3;

d) feeding into the second reaction mixture cyclohexanone oxime and a portion of the first reaction mixture, said cyclohexanone oxime being fed into the second reaction mixture through the second openings.

8. Process according to claim 7, wherein the process comprises

e) passing a third reaction mixture through the third tube, and keeping the third reaction mixture in circulation, said third reaction mixture comprising caprolactam, sulfuric acid and free SO_3;

9. Process according to claim 7 or claim 8, wherein the process is performed while a selected number of first channels and/or second channels and/or, optionally, third channels, is in closed position, said selected number being chosen such as to obtain a desired yield.

10. Process according to any one of claims 7 to 9, wherein the process is performed while a selected number of first channels and/or second channels, and/or, optionally third channels, is in closed position, said selected number being chosen such as to obtain a desired v_1/V_1 and v_2/V_2 and optionally v_3/V_3, wherein

v_1 = the velocity (in m/s) at which cyclohexanone oxime is fed through the first channels into the first reaction mixture,

V_1 = velocity of the first reaction mixture at the level of said first channels in the first tube, V_1 being defined as W1/A1, wherein W is the flow rate (in m^3/s) of the reaction mixture that is fed into the tube and A1 is the cross section area of the tube (in m^2) at the level where said first channels open into the tube.

v_2 = the velocity (in m/s) at which cyclohexanone oxime is fed through the second channels into the second reaction mixture,

V_2 = velocity of the second reaction mixture at the level of said second channels in the second tube, V_2 being defined as W2/A2, wherein W2 is the flow rate (in m^3/s) of the second reaction mixture that is fed into the second tube and A2 is the cross section area of the second tube (in m^2) at the level where said second channels open into the tube.

v_3 = the velocity (in m/s) at which cyclohexanone oxime is fed through the third channels into the third reaction mixture,

V_3 = velocity of the third reaction mixture at the level of said third channels in the third tube, V_3 being defined as W3/A3, wherein W3 is the flow rate (in m^3/s) of the third reaction mixture that is fed into the third tube and A3 is the cross section area of the third tube (in m^2) at the level where said third channels open into the tube.

11. Process according to any one of claim 10, wherein v_1/V_1 is smaller than v_2/V_2.

12. Process according to any one of claim 10, wherein v_1/V_1 is higher than v_2/V_2.

13. Process according to any one of claims 10 to 12, wherein v_1/V_1 is smaller than v_3/V_3.

14. Process according to any one of claims 10 to 12, wherein v_1/V_1 is higher than

v_3/V_3.

15. Process according to any one of claims 10 to 14, wherein v_2/V_2 is smaller than v_3/V_3.

16. Process according to any one of claims 10 to 14, wherein v_2/V_2 is higher than v_3/V_3.

ABSTRACT

The invention relates to an apparatus for preparing caprolactam, said apparatus comprising:

a) a first circulation system for keeping a first reaction mixture in circulation, said first circulation system comprising a first mixing device for admixing cyclohexanone oxime to the first reaction mixture, said first mixing device comprising:
   (a1) a first tube through which the first reaction mixture can flow;
   (a2) first channels disposed around the first tube through which first channels cyclohexanone oxime can be fed into the first reaction mixture, said first channels opening into the first tube; and
   (a3) one or more first closures, one or more of the first channels being closable with a first closure; and

b) a second circulation system for keeping a second reaction mixture in circulation, said second circulation system comprising a second mixing device for admixing cyclohexanone oxime to the second reaction mixture, said second mixing device comprising:
   (b1) a second tube through which the second reaction mixture can flow;
   (b2) second channels disposed around the second tube through which second channels cyclohexanone oxime can be fed into the second reaction mixture, said second channels opening into the second tube; and
   (b3) one or more second closures, one or more of the second channels being closable with a second closure.

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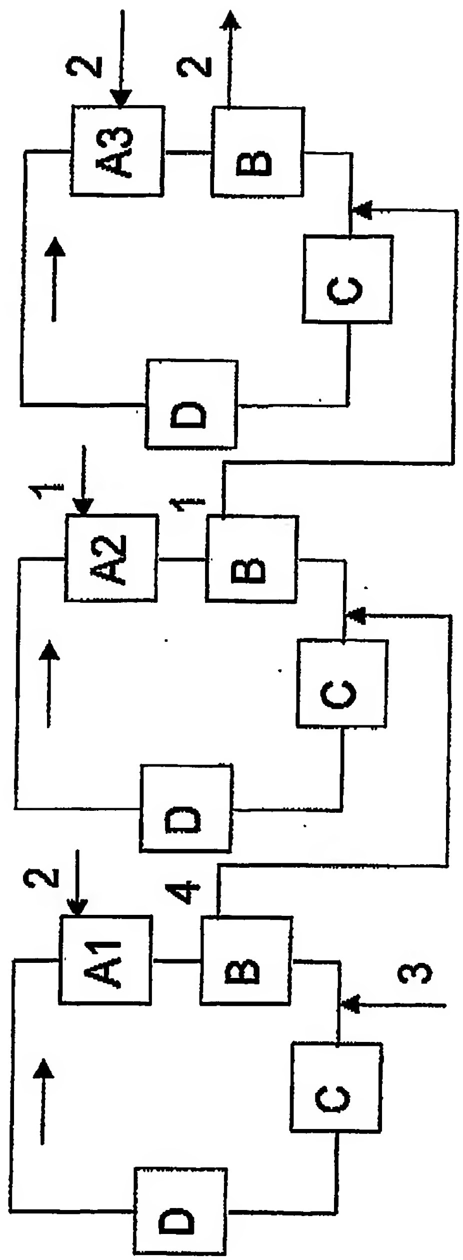

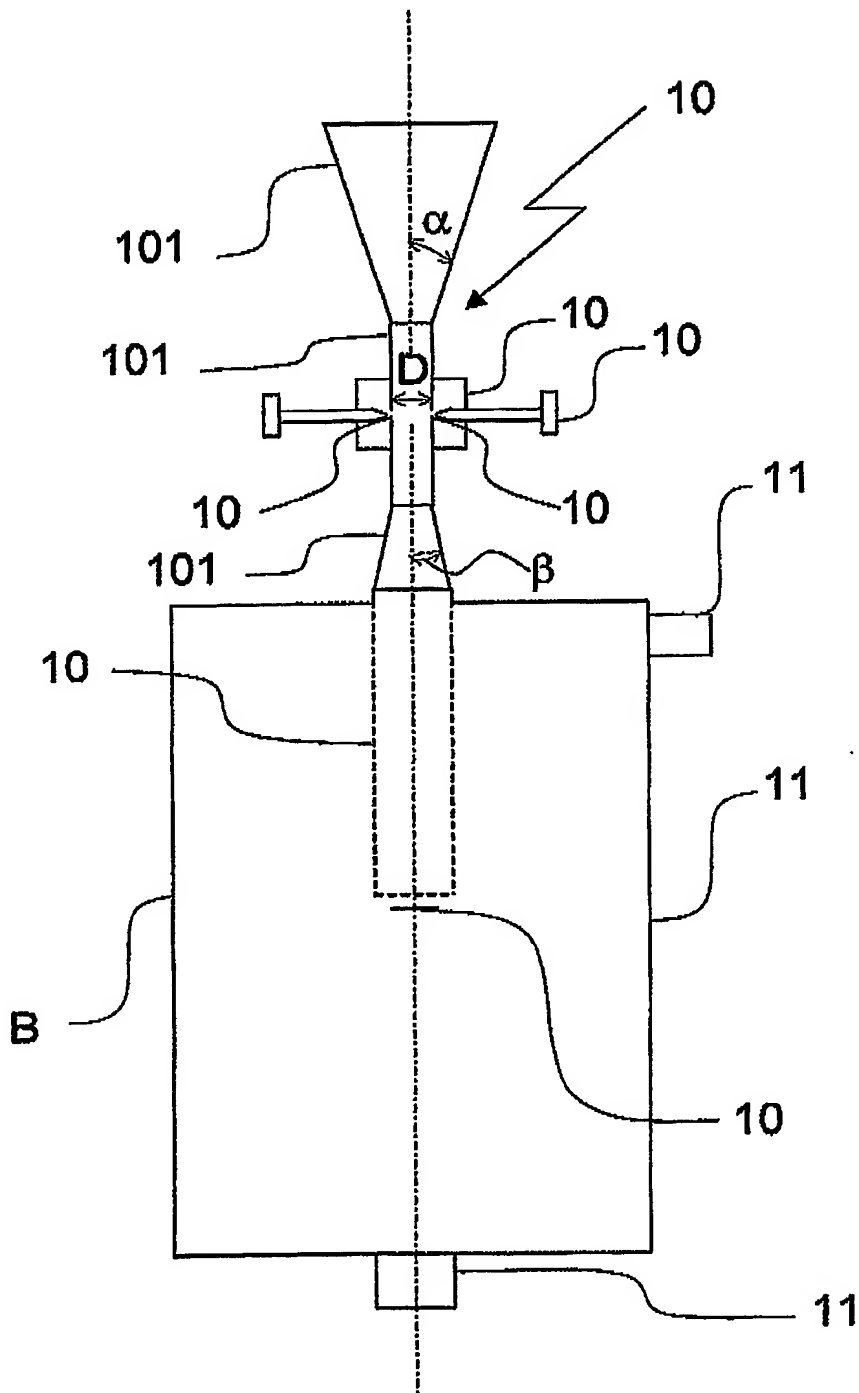
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